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Biofiltration of xylene emissions: bioreactor response to variations in the pollutant inlet concentration and gas flow rate

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Abstract

In order to remove xylene vapors from an air stream, an upflow laboratory scale biofilter was operated for a period of 2 months. The experimental study consisted of two different phases: in the first phase, the biofilter was operated at various gas flow rates and the xylene inlet concentration was maintained at 1.39 gm^{-3} . In the second phase, various inlet concentrations of the contaminant were tested at a constant gas flow rate of $0.4 \text{ m}^3 \text{ h}^{-1}$ corresponding to an empty bed residence time of 150 s. The biofilter response to steep and abrupt variations in the xylene inlet concentration and gas flow rate was examined. The results obtained revealed that the removal efficiency of the biofilter regained its high values (above 96%) in less than 24 h following the change to low concentrations and gas flow rate. Temperature measurements showed that the biofilter temperature strongly depends on the intensity of the microbial activity in the filter bed. The experimental mass ratio of carbon dioxide produced to the xylene removed was equal to 2.72 indicating that the contaminant was eliminated exclusively by aerobic biodegradation. These findings suggest that a follow up of the amount of carbon dioxide produced in the filter bed can be very helpful in monitoring the performance of the biofilter. For relatively small inlet loads of xylene, the contributions of the different sections of the biofilter to the removal efficiency of the contaminant and the carbon dioxide production were unevenly balanced but became more uniformly distributed for relatively high inlet loads.

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1. Introduction

The atmospheric emissions of volatile organic compounds (VOCs) represent a major contributor to the deterioration of air quality and pollution of the environment. Among these VOCs, xylene (or dimethylbenzene) is a hazardous chemical that can be found in many consumer products such as paints, lacquers, varnishes, adhesives, cements, inks and dyes, cleaners and degreasers, aviation gasoline, etc. It is also used in the manufacture of plastics and synthetic fibers, insecticides and pesticides, leather goods, and other chemicals. A considerable share (over 60%) of the total emissions of xylene into the atmosphere originates from industrial facilities. These industrial emissions are estimated at about 7300 metric tons per year in Canada and 34,000 metric tons per year in the United States [1]. In order to abide by the regulations of the local authorities and governments

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on the protection of the environment and air quality, industrial plants need to eliminate or reduce the content of toxic chemicals in gaseous effluents prior to their release into the atmosphere. In this context, biofiltration has emerged as an attractive process for the elimination of volatile organic compounds from waste air streams. The biofiltration process has various advantages compared to the conventional processes used in the treatment of gaseous effluents. It is an environment friendly technology appropriate for the treatment of dilute emissions. In addition to its low capital and operating cost [2,3], biofiltration is convenient for the processing of discontinuous emissions [4] and can be used to eliminate a variety of VOCs simultaneously [5–7].

In biofiltration, organic contaminants are degraded by aerobic heterotrophic microbial species. The polluted air stream is passed through the biofiltration column and flows through the packing material of the filter bed. The microbial population is immobilized on the surface of the packing material where they form an active biofilm layer. Because of the concentration gradient between the two phases, the contaminant diffuses from the gas phase into the liquid biofilm and is

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Nomenclature

$C_{\rm in}$	inlet concentration of xylene in the gas
	phase $(g m^{-3})$
$C_{\rm out}$	exit concentration of xylene in the gas
	phase $(g m^{-3})$
$C_{\rm CO_2,in}$	inlet concentration of carbon dioxide in the
-	gas phase $(g m^{-3})$
$C_{\rm CO_2,out}$	exit concentration of carbon dioxide in the
	gas phase $(g m^{-3})$
ΔCO_2	carbon dioxide production $(g m^{-3})$
EC	elimination capacity of xylene $(g m^{-3} h^{-1})$
IL	inlet load of xylene $(g m^{-3} h^{-1})$
$P_{\rm CO_2}$	carbon dioxide production rate $(g m^{-3} h^{-1})$
\bar{Q}	gas flow rate $(m^3 h^{-1})$
T	temperature of the biofilter (°C)
V	volume of the filter bed (m^3)
X	removal efficiency of xylene (dimensionless)
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then biodegraded by the microorganisms present in the liquid biolayer. These microorganisms ensure their growth and survival using the carbon source from the organic pollutant and the nutrients available in the packing material. Nonetheless, the filter bed is usually irrigated by a nutrient solution that provides a supplement of nutrients to the microorganisms and improves their microbial activity.

The performance of a biofiltration unit depends on a large number of factors. Among these factors are the biodegradation effectiveness and selectivity of the microbial population, composition of the waste gas stream, degree of recalcitrance of the contaminants, structure and composition of the packing material, operating conditions of the biofilter, the bioreactor design, etc. Accordingly, it is crucial to better understand the various aspects of the process so as to improve the efficiency of biofilters. In this framework, numerous studies investigated the biofiltration of gaseous effluents containing xylene either as a sole contaminant [8–10] or in a mixture with other aromatic compounds [9,11–18]. In the present study, the biofilter response to steep and abrupt variations in the xylene inlet concentration and gas flow rate is examined using a laboratory scale upflow biofiltration unit. The interdependence of the biofilter performance and both the filter bed temperature and carbon dioxide production are investigated as well.

2. Materials and methods

Experiments were conducted over a period of 2 months using the biofiltration unit as shown in Fig. 1. This unit consists mainly of a biofiltration column, a humidification tower, several instrumentation devices, and accessory equipment. The Plexiglas biofilter unit is a 1.5 m high cylinder with an inner diameter of 0.15 m. It is subdivided into three separate identical sections and packed with conditioned spherical peat particles [19] over a height of 0.33 m in each section. The packing material was manufactured in our laboratory using an agglomerating apparatus and consisted of 70% (v/v) peat and 30% (v/v) of a certain mineral additive with good bind-



Fig. 1. Experimental setup.

ing capacity. The filter bed was initially inoculated with a specific microbial activated consortium (EVB110) provided by GSI Environnement Inc. (Sherbrooke, Oue., Canada). The consortium consisted essentially of specific microbial aerobic and facultative anaerobic species. A xylene contaminated air stream is generated by mixing a primary stream of humidified clean air and a secondary stream of air saturated with xylene (Fig. 1). In order to avoid dryness of the packing material in the filter bed, the primary stream flows through a humidification tower and reaches 95% relative humidity prior to mixing. The flow rates of both primary and secondary streams are controlled using previously calibrated flowmeters to obtain the desired gas flow rate and concentration of the contaminant at the biofilter entrance. The polluted air stream is injected at the base of the biofiltration column and flows through the interstices of the filter bed where biodegradation of the contaminant takes place. The biofilter is occasionally irrigated by a nutrient solution that maintains adequate moisture of the filter bed and supplies the microorganisms with additional nutrients [20]. The bed was intermittently irrigated with the nutrient solution, the composition of which is as follows for 11 of distilled water: KH₂PO₄, 2.81 g; NH₄Cl, 1.87 g; NH₄HCO₃, 3.75 g; NH₄SO₄, 1.87 g; and some traces of Co, Cu, Fe, Mn, Na, Mo, and Zn.

In order to examine the performance of the biofilter, inlet and outlet concentrations of xylene and carbon dioxide were measured daily. Air samples were withdrawn from the biofiltration column and directed to a total hydrocarbon analyzer (model FIA-220, Horiba) and a carbon dioxide analyzer (Ultramat 22P, Siemens) for xylene and CO₂ concentration measurements, respectively. The gas analysis equipment was calibrated daily prior to these measurements. Clean air and air prepared with various known xylene and carbon dioxide concentrations served as the calibration standards. The temperature of the filtering medium was recorded daily at three different levels of the biofiltration column using T type thermocouples.

The experimental results will be expressed in terms of the xylene inlet load, IL (g m⁻³ h⁻¹); the elimination capacity, EC (g m⁻³ h⁻¹); the biofilter removal efficiency, *X* (%); the carbon dioxide production, ΔCO_2 (g m⁻³); and the carbon dioxide production rate, P_{CO_2} (g m⁻³ h⁻¹). These quantities are defined by

$$IL = \frac{QC_{in}}{V}$$
(1)

$$EC = \frac{Q(C_{\rm in} - C_{\rm out})}{V}$$
(2)

$$X = \frac{C_{\rm in} - C_{\rm out}}{C_{\rm in}} \times 100 \tag{3}$$

$$\Delta \text{CO}_2 = C_{\text{CO}_2,\text{out}} - C_{\text{CO}_2,\text{in}} \tag{4}$$

$$P_{\rm CO_2} = \frac{Q(C_{\rm CO_2,out} - C_{\rm CO_2,in})}{V}$$
(5)

Here, C_{in} and C_{out} (g m⁻³) represent the respective inlet and exit xylene concentration in the gas phase, Q (m³ h⁻¹) the volumetric airflow rate, and V (m³) the volume of the filter bed. The carbon dioxide concentrations measured at the inlet and exit of the biofilter are denoted by $C_{CO_2,in}$ and $C_{CO_2,out}$ (g m⁻³), respectively.

3. Results and discussion

During the first month of the experimental study, the air stream was injected at the biofilter entrance with a constant xylene inlet concentration of $1.39 \,\mathrm{g}\,\mathrm{m}^{-3}$. The gas flow rate was varied between 0.4 and $1.1 \text{ m}^3 \text{ h}^{-1}$ corresponding to an EBRT varying between 150 and 56 s, corresponding to inlet loads between 34 and 95 g m⁻³ h⁻¹, and the biofilter response to these variations was examined. Fig. 2 shows the gas flow rate variations along with the removal efficiency of xylene recorded during the same period of experimentation. Initially, the gas flow rate Q was set at its lowest value of $0.4 \text{ m}^3 \text{ h}^{-1}$. After 5 days of operation, Q was abruptly increased to higher values as seen in Fig. 2. Later, the gas flow rate was subject to a steep decrease back to its initial value. As one can see, the removal efficiency of the contaminant X diminishes when the operating gas flow rate is increased but X becomes much larger when O decreases. This can be attributed to the fact that for higher gas flow rates, the gas residence time in the biofiltration column is smaller. Consequently, the contact time between the polluted air stream and the degrading microorganisms is smaller and the biodegradation ability of the filter bed diminishes leading to lower values of the removal efficiency of the contaminant. Moreover, it is important to notice that despite the precipitous variations in the gas flow rate, the removal efficiency of the biofilter regained its original values that exceed 96% in less than 24 h when Q was reset to $0.4 \text{ m}^3 \text{ h}^{-1}$ (Fig. 2).

In the biofiltration process, xylene is biodegraded under aerobic conditions to carbon dioxide, water, and biomass. Hence, monitoring the carbon dioxide concentration in the gas phase can provide valuable information on the operation of the biofilter. The daily measurements of the carbon dioxide production are presented in Fig. 3 along with the removal efficiency of the biofilter. The measurements in Fig. 3 show that an increase/decrease in the removal efficiency is in general accompanied with an increase/decrease in the carbon dioxide production in the filter bed. Furthermore, the variation trend of both X and CO₂ produced are reminiscent, which confirms the interdependence of these two parameters. Fig. 4 shows a plot of the filter bed performance versus the amount of carbon dioxide produced. As the removal efficiency increased from 48 to 99%, the production of CO₂ increased from 1.98 to 4.68 g m^{-3} .

In the second month of experimentation, the biofiltration column was fed with an air stream at a constant gas flow rate of $0.4 \text{ m}^3 \text{ h}^{-1}$ corresponding to an EBRT varying between 150 and 56 s, and the biofilter response to variations in the



Fig. 2. Removal efficiency of xylene and gas flow rate vs. time for $C_{\rm in} = 1.39 \, {\rm g \, m^{-3}}$.

inlet concentration of xylene was examined. The biofilter was subject to steep variations in the concentration $C_{\rm in}$ that ranged between 0.48 and 2.69 g m⁻³, corresponding to inlet loads varying between 12 and 67 g m⁻³ h⁻¹. The daily mea-

surements of the contaminant inlet load, IL; the elimination capacity, EC; and the biofilter removal efficiency, *X* are presented in Fig. 5. These measurements show that the removal efficiency exceeded 98% for inlet loads up to $36 \text{ g m}^{-3} \text{ h}^{-1}$



Fig. 3. Removal efficiency of xylene and carbon dioxide production vs. time.



Fig. 4. Removal efficiency of xylene vs. carbon dioxide production in the biofilter.



Fig. 5. Inlet load, elimination capacity, and removal efficiency of xylene vs. time for $Q = 0.4 \text{ m}^3 \text{ h}^{-1}$.



Fig. 6. Elimination capacity of xylene and the filter bed average temperature vs. time.

and varied between 73 and 93% for higher inlet loads. Nevertheless, given the degree of recalcitrance of the contaminant, the performance of the biofilter was good over the total range of inlet concentrations tested. In addition, in spite of the abrupt variations in the inlet load of xylene, the removal efficiency of the biofilter regained its high values that exceed 98% in less than 24 h, following the change to low concentrations (Fig. 5).

The filter bed temperature was monitored at three different levels of the biofiltration column. The daily measurements of the average temperature T are shown in Fig. 6 along with the elimination capacity EC of the biofilter. As one can see, when the temperature rises or diminishes, the elimination capacity tends to follow the same trend of variation. In fact, the reaction of biodegradation of xylene in the wet biofilm is exothermic. Thus, an increase in the intensity of the biodegradation activity gives rise to a higher yield in the elimination of the contaminant and an augmentation of the filter bed temperature, simultaneously. In Fig. 7, average temperature of the biofilter is plotted versus the elimination capacity. The plot shows that T is an increasing function of EC, and as the temperature is increased from 26 to 28 °C, the elimination capacity increases from 12 to $61 \text{ g m}^{-3} \text{ h}^{-1}$. Hence, the intensity of the microbial activity in the filter bed strongly depends on the biofilter temperature.

In Fig. 8, the biofilter elimination capacity measurements are presented along with the carbon dioxide production. In general, an increase/decrease in the elimination capacity is accompanied with an increase/decrease in the production of CO₂, a behavior similar to the one observed with the temperature (Fig. 6). For a more quantitative analysis of these results, the carbon dioxide production rate, P_{CO_2} is plotted against the biofilter elimination capacity, EC (Fig. 9). Experimental data reveal that the variation of P_{CO_2} versus EC is sensibly linear. The average ratio of the measured P_{CO_2} to EC was equal to 2.72 with a standard deviation of 0.29. In other words, the mass of carbon dioxide produced per mass of xylene eliminated was approximately equal to 2.72. This experimental value is lower than 3.3, the stoichiometric ratio in the case of complete chemical oxidation of the contaminant. The discrepancy between these two ratios is typical since during the process of biodegradation of the organic pollutant, some of the removed carbon is converted into biomass for microbial growth. Moreover, a portion of the carbon dioxide produced may partly accumulate in the wet biofilm as one of its solute species [9]. Nonetheless, the relatively small difference between the complete chemical oxidation based ratio and the experimental ratio indicates that removed xylene is eliminated by biodegradation rather than by any other physical or chemical process such as adsorption. In conclusion, a follow up of the amount of carbon dioxide produced in the filter bed can be very helpful in monitoring the biofilter performance.

As mentioned earlier, the biofiltration column is subdivided into three identical sections operating in sequence (Fig. 1). In order to analyze the composition of the gas phase, samples were withdrawn from the inlet and outlet of the biofilter and the exit of the first two sections as well. As



Fig. 7. Filter bed average temperature vs. elimination capacity of xylene.



Fig. 8. Elimination capacity of xylene and carbon dioxide production vs. time.



Fig. 9. Carbon dioxide production rate vs. elimination capacity of xylene.

a result, the contribution of the three different sections of the bioreactor to its overall performance can be compared to each other. While the outcome of such comparison may be less relevant than the overall efficiency of the biofilter, it may provide further insight on the operation of the biofiltration column. Fig. 10 shows the removal efficiency of the three sections of the biofilter for various inlet loads of xylene. Here, the removal efficiency of an individual section is defined as the ratio of the difference between the inlet and exit concentration of xylene in this section to the concentration of xylene C_{in} at the entrance of the biofilration column. Thus, the summation of the three individual section efficiencies is equivalent to the overall removal efficiency X of the biofilter given by Eq. (3). Experimental data revealed that for relatively small inlet loads, the contributions of the different sections of the biofilter to the biodegradation of xylene were not evenly balanced. For instance, at an inlet load of $12.8 \,\mathrm{g}\,\mathrm{m}^{-3}\,\mathrm{h}^{-1}$, the removal efficiency in the first section was equal to 71.7% compared to 23.2 and 3.5% in the second and third sections, respectively. In this case, a majority of the contaminant was eliminated in the first section. However, as IL increased, the removal efficiency in the lower section diminished and the biodegradation of xylene became more uniformly distributed among the three sections of the bioreactor (Fig. 10). This behavior was anticipated since at relatively low inlet loads, the amount of xylene available for biodegradation is small enough so that a large portion of the pollutant is biodegraded in the lower

section of the biofilter. In this situation, only a small portion of xylene is available for elimination in the upper sections and the biofilter overall removal efficiency is close to 100%. On the other hand, for relatively high inlet loads of the pollutant, each section eliminates as much contaminant as possible and a complete removal of xylene in the biofilter is not achieved. Because of the uniformity of the spatial distribution of the degrading microorganisms in the filter bed, each section contributes equally to the overall removal efficiency obtained. In the same way, one can analyze the measurements of carbon dioxide concentration in different sections of the biofiltration column. Fig. 11 shows the CO₂ production percentage at each section of the biofilter versus the elimination capacity of xylene. Here, the carbon dioxide production percentage for an individual section is defined as the ratio of the CO₂ produced in this section to the total production of CO_2 in the bioreactor. Once again, when operating at a relatively small inlet load of xylene, a larger percentage of the carbon dioxide produced was generated in the lower section of the biofiltration column. Furthermore, the spatial distribution of the CO_2 production in the biofilter became more uniform among the different sections as IL increased (Fig. 11). This behavior is similar to the one observed previously for the removal efficiency of the individual sections, and such similarity was anticipated since the carbon dioxide production is strongly associated with the intensity of the microbial activity in the filter bed.



Fig. 10. Removal efficiency in different sections of the biofilter vs. inlet load of xylene.



Fig. 11. Carbon dioxide production percentage in different sections of the biofilter vs. elimination capacity of xylene.

4. Conclusion

An experimental study on the removal of xylene vapors from an air stream was conducted on an upflow laboratory scale biofilter over a period of 2 months. In the first phase of this study, the biofiltration column was fed with an air stream characterized with a constant xylene inlet concentration of 1.39 g m^{-3} , and the empty bed residence time of the injected air stream was varied between 150 and 56 s. In the second phase, the biofilter was operated under a constant empty bed residence time of 150 s but various inlet concentrations of the contaminant were tested $0.48-2.69 \text{ g m}^{-3}$. During the period of experimentation, the biofilter response to steep and abrupt variations in the xylene inlet concentration and gas flow rate was investigated. The results obtained showed that the removal efficiency of the bioreactor regained its high values (above 96%) in less than 24 h following the change to low concentrations and gas flow rate.

Temperature measurements revealed that the biofilter temperature strongly depends on the intensity of the microbial activity in the filter bed. Increase from 26 to 28 °C in the average temperature of the filter bed was accompanied with an increase from 12 to 61 g m⁻³ h⁻¹ in the elimination capacity.

The carbon dioxide concentration in the gas phase was measured at the inlet and exit of the biofiltration column. As the removal efficiency of the biofilter increased from 48 to 99%, the amount of CO_2 produced increased from 2.02 to 7.6 g m⁻³. Furthermore, the mass of carbon dioxide produced per mass of xylene eliminated was equal to 2.72. These findings suggest that the removed xylene was eliminated exclusively by aerobic biodegradation, and that a follow up of the amount of CO_2 produced in the filter bed can be very helpful in monitoring the performance of the biofilter.

For relatively small inlet loads of xylene, the contributions of the different sections of the biofilter to the removal efficiency of the contaminant and the carbon dioxide production were unevenly balanced but became more uniformly distributed for relatively high inlet loads.

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